

Variability of ozone loss during Arctic winter (1991 to 2000) estimated from UARS Microwave Limb Sounder measurements

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Abstract. A comprehensive analysis of version 5 Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS) ozone data using a Lagrangian Transport (LT) model provides estimates of chemical ozone depletion for the 1991-1992 through 1997-1998 Arctic winters. These new estimates give a consistent, three-dimensional picture of ozone loss during seven Arctic winters; previous Arctic ozone loss estimates from MLS were based on various earlier data versions and were done only for late winter and only for a subset of the years observed by MLS. We find large interannual variability in the amount, timing, and patterns of ozone depletion and in the degree to which chemical loss is masked by dynamical processes. Analyses of long-lived trace gas data suggest that the LT model sometimes overestimates descent at levels above ~ 520 K, so we have most confidence in the results at lower levels. When the vortex is shifted off the pole and the cold region is near the vortex edge (e.g., late winter 1993 and 1996), most rapid ozone depletion occurs near the vortex edge; when the vortex and cold region are pole-centered (e.g., late winter 1994 and 1997), most ozone loss takes place in the vortex core. MLS observed the most severe ozone depletion in 1995-1996, with about 1.3 ppmv cumulative loss for the winter at 465 K by 3 March 1996; ~ 1.0 ppmv cumulative loss is seen at 465 K by mid-March 1993. Analyses of MLS data show significant ozone loss during January in most years, ranging from ~ 0.3 to 0.6 ppmv at 465 K. A modified LT model used with the limited MLS data in 2000 gives rough estimates of ~ 0.04 ppmv/day and 0.006-0.012 ppmv/day during 2-12 February and 12 February-29 March 2000, respectively, broadly consistent with other studies of the 1999-2000 winter. Estimates of depletion in MLS column ozone above 100 hPa are considerably smaller than other reported column loss estimates, primarily because many estimates include loss below 100 hPa and because MLS does not continuously observe the Arctic after early spring. Our results from analyses of MLS data confirm previous conclusions of broad overall agreement between many ozone loss estimates in the Arctic lower stratosphere near 450-480 K.

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1. Introduction

Measurements from the Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS) have been instrumental in studying Arctic ozone loss. *Manney et al.* [1994b] first presented conclusive evidence of widespread Arctic ozone loss using MLS and UARS Cryogenic Limb

Array Etalon Spectrometer (CLAES) data; they showed that the decrease in MLS-observed ozone in the lower stratosphere in February and March 1993 was inconsistent with the coincident time-evolution of long-lived trace gases from CLAES, indicating that dynamical processes alone could not be responsible for the observed ozone decrease. These results confirmed that substantial ozone loss can and does occur throughout the Arctic lower stratospheric vortex.

Attempts to quantify the ozone loss observed by MLS have been made in two ways. MacKenzie *et al.* [1996] used observed MLS ClO data and a chemical model to estimate the expected ozone change due to chemistry, and compared this with the observed MLS ozone values for the 1992-93 northern hemisphere (NH) winter. Manney *et al.* [1995b, c] used a Lagrangian transport (LT) model, transporting trace gases passively along calculated parcel trajectories, to quantify the changes expected from dynamical processes in the 1992-93 NH winter. The difference between these changes and the observed ozone changes gives an estimate of chemical loss. The results of Manney *et al.* [1995b] and MacKenzie *et al.* [1996] were broadly consistent and indicated chemical ozone loss of about 1%/day in February/March 1993 near 465 K, with 20-30% of that loss masked by transport.

Manney *et al.* [1995b] were the first to use the general method of comparing modeled passively-transported ozone to observations to quantify ozone loss. This method has since been widely used with a variety of datasets and transport models to quantify Arctic ozone loss [e.g., Manney *et al.*, 1997; Deniel *et al.*, 1998; Goutail *et al.*, 1999; Guirlet *et al.*, 2000; Sinnhuber *et al.*, 2000; Hoppel *et al.*, 2002, and references therein]. Limited comparisons of some of these transport-model studies, and studies using Match [e.g., Rex *et al.*, 1997, 2002, and references therein], vortex-averaged [e.g., Knudsen *et al.*, 1998, and references therein], and trace-gas correlation methods [e.g., Müller *et al.*, 1997a, b, and references therein] have been done by Harris *et al.* [2002], who found broadly consistent results between various methods.

Manney *et al.* [1995c] used CLAES long-lived trace gas data to evaluate the model's ability to simulate transport. Manney *et al.* [1995a, 1996a, b, 1997] used a revised LT model (based on reverse trajectory calculations rather than forward trajectory calculations with regridding) to study Arctic ozone loss in February-early March 1994, January and February 1995 and 1996, and February-early April 1997. MLS also observed ozone during the 1991-1992 Arctic winter [e.g., Waters *et al.*, 1993] and parts of the 1997-1998 winter [e.g., Orsolini *et al.*, 2001], periods for which similar calculations have not been published. MLS also made a limited number of observations during a two-week period in February 2000 and three days in late March 2000 [Santee

et al., 2000].

The calculations described above were done using version 3 (V3) or version 4 (V4) (for Manney *et al.* [1996b, 1997]) MLS data. Manney *et al.* [1995c] used version 7 CLAES CH₄ and N₂O data and V3 MLS H₂O for long-lived trace gas calculations. The current versions (considered definitive) of MLS and CLAES data are version 5 (V5) and version 9 (V9), respectively. V5 MLS data represent major improvements in the retrieval software [Livesey *et al.*, 2002], and changes between versions result in differences in estimates of ozone loss. The calculations previously reported by Manney *et al.* [1995a, b, 1996a, b, 1997] not only were done with different data versions, but also did not include several winters observed by MLS. In addition, most of them covered only the late winter (February/March) periods observed by MLS. Several studies [e.g., Manney *et al.*, 1996a, b; Rex *et al.*, 1998, 2002] suggest significant Arctic ozone loss during January and this loss has not been well-reproduced by models [e.g., Becker *et al.*, 2000, and references therein].

To get a comprehensive and consistent view of the amount of Arctic ozone depletion during the UARS MLS mission, we have extended previous LT calculations to provide ozone loss estimates in each of the winters observed by MLS. These calculations are done using V5 MLS data and the same LT model version, so interannual comparisons can be made. Estimates of cumulative ozone loss over the 1991-1992 through 1997-1998 Arctic winter periods observed by MLS and estimates of loss in column ozone above 100 hPa are presented. These results not only allow us to show interannual variability in ozone loss, but also facilitate comparison of MLS results with other Arctic ozone loss studies. We also include modified LT calculations to estimate ozone loss from the limited MLS data in 1999-2000.

2. Data and Analysis

2.1. Instruments and Coverage

The UARS MLS instrument [Barath *et al.*, 1993] measured ozone at 205 GHz in the stratosphere and lower mesosphere beginning in September 1991. Figure 1 shows MLS measurement coverage during the NH winter. The UARS platform orientation causes the MLS instrument to switch between viewing ~34°S to 80°N and ~80°S to 34°N approximately every 36 days [e.g., Reber, 1993]. This provides coverage of high northern latitudes in December and February, with early winter observations beginning from early December to mid-November, and late winter observations extending to early or middle March; UARS yaws occurred a few days earlier in each succeeding year. MLS took continuous measurements during the north-looking periods in 1991-1992 and 1992-1993. In February/March 1994, MLS

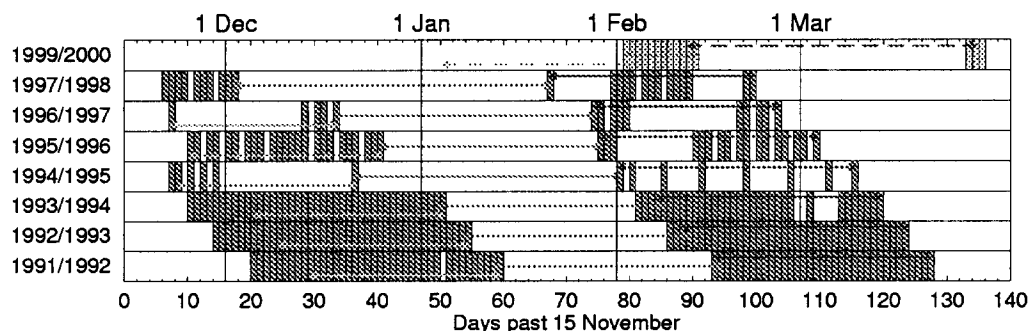


Figure 1. MLS ozone measurement coverage during the Arctic winter. No measurements were taken in the 1998–1999 winter. Thick grey-shaded bars show MLS north-looking observations; lighter shading in 2000 indicates that those measurements were geographically limited. Thin lines indicate time periods of Lagrangian Transport (LT) model runs discussed below; blue lines are “early winter” runs, green lines “mid-winter” runs, and red lines “late winter” runs; lines for 2000 are dashed to indicate the different initialization method for the LT model in that year.

measurement coverage was affected by problems with the antenna scanning mechanism; in the following winters, MLS normal scanning operations were attempted every other day, or two out of three days, to conserve instrument lifetime [e.g., Livesey et al., 2002]. During the 1994–1995 winter, scanning problems were severe, allowing only a few days of normal observations. Measurements in the 1997–1998 winter were obtained after deactivation of the 63-GHz radiometer, which provided pointing information. Largest differences between retrievals for this operating mode and “standard” retrievals are at 46 hPa and higher pressures; even here, the two types of retrievals are consistent to within $\sim 10\%$ [Livesey et al., 2002]. After early 1998, MLS was placed in standby mode; it was turned on for limited Arctic observations from 2–12 February and 27–30 March 2000. The 2000 Arctic data come from scanning observations only at high northern latitudes and, during February, only on the “day-time” side of the orbit. While MLS performed flawlessly during this period, there were unfortunately problems transmitting the data from the satellite; thus the 2000 observations have limited geographic coverage [Santee et al., 2000].

The UARS CLAES instrument provided data, including several long-lived trace gases, with the same spatial coverage as MLS from October 1991 through April 1993, when the supply of cryogen for the planned 18-month mission was depleted [e.g., Roche et al., 1993]. The north-looking periods covered by the CLAES data are two or three days shorter at each end than those shown in Figure 1 because the CLAES door had to be closed during each yaw maneuver.

2.2. Data Versions

Characteristics and validation of V3 ozone data are described by Froidevaux et al. [1996]. V5 data are retrieved on pressure surfaces with ~ 2.5 km spacing (six levels per decade in pressure), twice as fine a grid as previous versions. The vertical resolution of MLS V5 ozone is ~ 3.5 – 4 km in the lower stratosphere; precision is ~ 0.25 – 0.3 ppmv at and above 68 hPa, and ~ 0.4 ppmv at 100 hPa. Estimated accuracy is 6% at 46 hPa and above, and the maximum of 0.25 ppmv (0.1 ppmv) or 15% at 68 (100) hPa. Details of differences between V4 and V5 MLS ozone are given by Livesey et al. [2002]; they found that V5 ozone data agree more closely with correlative data in the lower stratosphere than did previous data versions. Figure 2 shows V4/V5 differences as a function of equivalent latitude (EqL)¹ and potential temperature (θ) on 11 February 1993, the initialization date of one of the LT model runs discussed below. Significant differences in the vertical ozone gradients in the polar and midlatitude lower stratosphere are apparent in the changing differences as a function of height. Changes in the vertical ozone gradients result in significant changes in the passive ozone calculated by the LT model, as different amounts of ozone are transported down by diabatic descent. Figure 3 shows the time evolution of vortex-averaged V4 and V5 MLS ozone in February/March 1993; in addition to differences in the vertical ozone gradients shown above, changes in the time evolution, similar in character to changes reported by Livesey et al. [2002] for the south-

¹The latitude that would enclose the same area between it and the pole as a given potential vorticity (PV) contour, e.g., Butchart and Remsberg [1986]

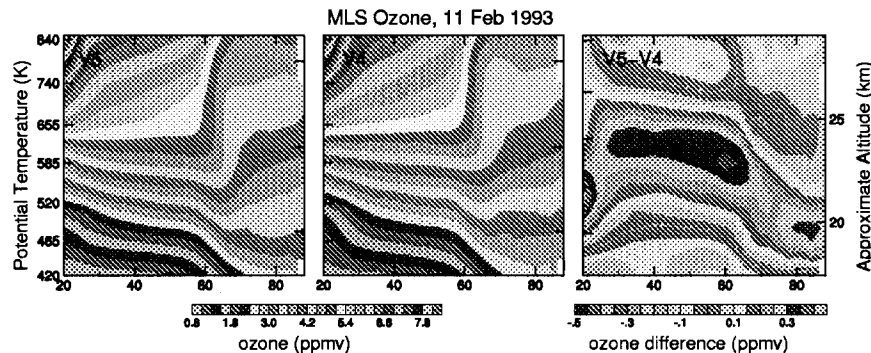


Figure 2. MLS ozone on 11 February 1993, as a function of equivalent latitude (EqL) and potential temperature (θ): (a) Version 5 (V5), (b) Version 4 (V4), and (c) V5–V4 differences (white line is zero contour).

ern hemisphere, are apparent between the two data versions. With changes in the vertical gradients affecting the modelled ozone, and changes in observed time evolution, significant differences are seen between estimates of ozone loss from different MLS data versions.

V9 CLAES N_2O and CH_4 data, and MLS H_2O data from “prototype V104” retrievals [Pumphrey, 1999], are also compared with LT model results. Roche et al. [1996] discussion validation and quality of V7 CLAES N_2O and CH_4 data. Numerous improvements were made in the retrieval software in version 8, which are reflected in the V9 data. Both N_2O and CH_4 agree better with correlative data in V9 than in V7. The vertical resolution of the CLAES data is ~ 3 km, and the data are reliable down to ~ 450 K in the winter vortex region (and somewhat higher at lower latitudes and/or higher temperatures when a given isentrope corresponds to a higher pressure).

2.3. Analysis and Modeling Methods

MLS and CLAES data are gridded by partitioning all data from a 24-hour period into 4° latitude by 5° longitude bins and assigning to each gridpoint a weighted average of the values within the bin. UARS data are interpolated to isentropic surfaces using temperatures from the U.K. Met Office analyses [Swinbank and O’Neill, 1994]. PV is also calculated from the Met Office data and used to define the vortex for averaging and to map UARS and LT model data as a function of EqL and θ . Using a vortex-centered coordinate is helpful for Arctic ozone studies, facilitating detection of significant ozone changes in the highly asymmetrical and variable Arctic vortex that may be masked in zonal means [e.g., Manney et al., 1994b]. Figure 4 shows ozone changes during the late winter periods observed by MLS in 1992 through 1998, as a function of EqL and θ (a similar plot for February and March 2000 is shown by Santee et al. [2000]). The ob-

served changes suggest varying amounts of ozone loss during these winters, with some ozone decrease in the vortex region in all years except 1992 and 1998. To quantify the amount of chemical loss, though, we need an estimate of how ozone would have changed via transport processes in the absence of chemical processes.

The LT model [Manney et al., 1995a, b, c] uses a reverse trajectory procedure to transport parcels back from the gridpoints on each modeled day to the initialization day; trace gas values at the gridpoints are then taken to be the values interpolated to the parcel positions from observed trace gas fields on the initialization day. The calculations are done on eight isentropic surfaces: 385, 420, 465, 520, 585, 655, 740 and 840 K (covering ~ 15 to 30 km altitude), on a 4° latitude by 5° longitude grid from 0° to 80°N . The LT model is based on the trajectory code described by, e.g., Manney et al. [1994a]; Met Office winds and temperatures and the MIDRAD [Shine, 1987] radiation code are used to calculate the trajectories. The LT model was run for each late winter period observed by MLS, and each early winter period except 1997 when there were only a few days of data in December (Figure 1). The model was also run from the end of each early winter north-viewing period to the beginning of the following, late winter, north-viewing period (Figure 1). “Extended” runs were done by initializing LT model runs for the late winter periods with the results from the last day of the runs that covered the gap in MLS NH data; this is preferable to running the LT model for the combined period because it reduces the accumulated errors in parcel positions that grow over long trajectory runs. In most years there was little or no ozone loss before the beginning of these extended runs (see section 4 below); thus they provide an estimate of the cumulative chemical ozone loss through the time of the final north-looking MLS observations in each winter.

Column ozone above 100 hPa (Col100) is calculated from MLS data by integrating the retrieved abundances over the

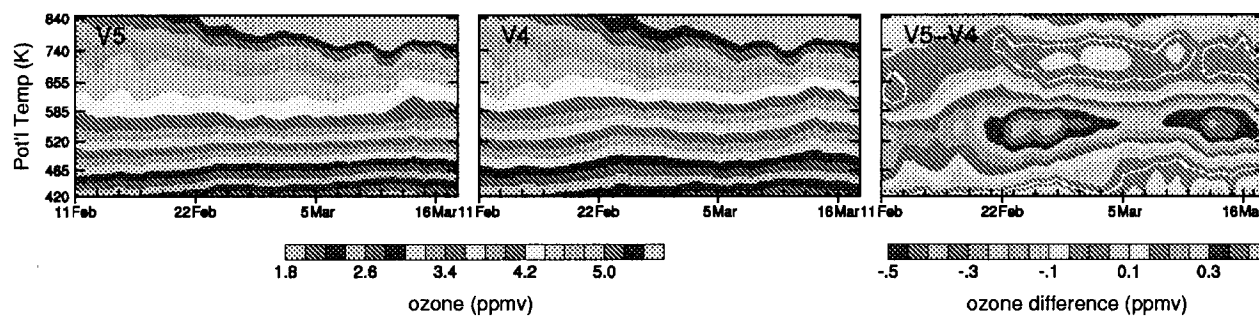


Figure 3. MLS ozone averaged in the vortex, as a function of θ and time in February and March 1993: (a) V5, (b) V4, and (c) V5–V4 differences (white line is zero contour).

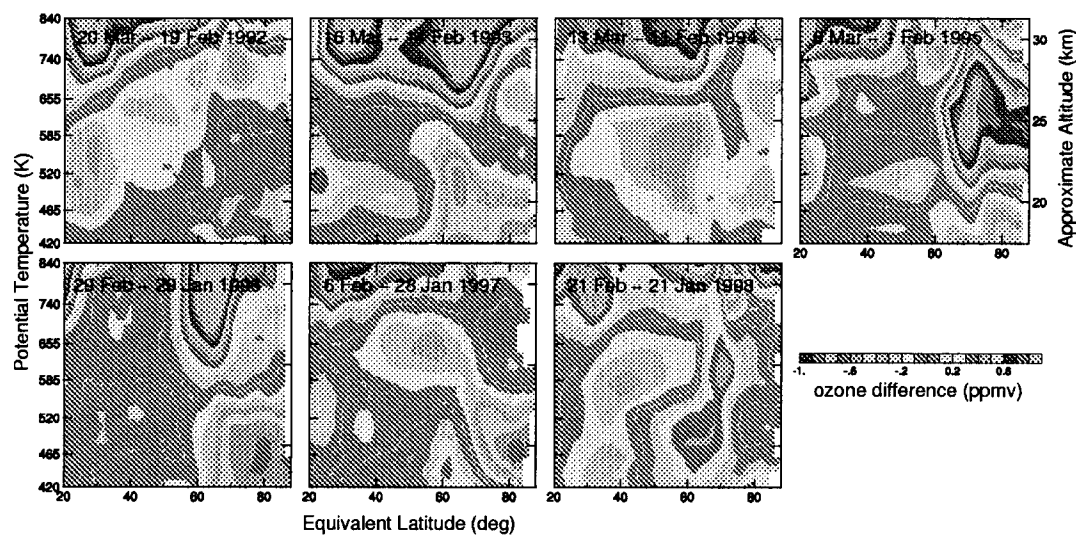


Figure 4. MLS observed ozone changes (ppmv) as a function of EqL and θ , from the beginning to the end of the seven late winter periods observed by MLS. White line is zero contour.

MLS retrieval grid (6 points per decade change in pressure), following the assumption of triangular basis functions for the profile representation (linear change in mixing ratio versus $\log(\text{pressure})$). In the polar winter, the tropopause pressure may range from ~ 120 to ~ 300 hPa, so Col100 represents most, but not all, of the stratospheric column. Col100 is calculated from LT model results using the same algorithm, after some manipulation of the LT fields. Since little or no ozone loss is expected above ~ 30 hPa, and since the lack of a parameterization of chemical production in the midstratosphere affects the LT calculations above this level, LT ozone is relaxed to MLS observations over the three levels from 22 to 10 hPa after interpolation to pressure surfaces. If LT ozone is missing on the 100-hPa surface, it is filled, if possible, by extrapolation from the two levels above. Gaps of one or two gridpoints are filled with a weighted average of surrounding points. If, after these procedures, good data are not present at all levels from 100–22 hPa, a bad data value is returned for Col100 for that gridpoint.

3. Spatial development of ozone loss

We show below an overview of patterns and variability in ozone loss in years with most complete coverage. In order to detect chemical ozone changes, it is necessary to first understand how well the LT model simulates changes from transport. Manney *et al.* [1995c] used CLAES N_2O and CH_4 and MLS H_2O to examine the accuracy of the transport model in the 1992–1993 winter; they found an overestimate of diabatic descent in 1993 at levels above ~ 520 K. Figure 5 shows vortex-averaged V9 N_2O from CLAES and from the LT model initialized with V9 CLAES data for the February/March 1992 and 1993 periods. The difference plots suggest an overestimate of descent in the vortex above ~ 520 K in both 1992 and 1993, and an underestimate at lower levels. All of the long-lived trace gas data used here become increasingly uncertain at levels below about 500 K, especially toward the end of these periods, when increasing temperatures mean that the measurements on a given isentrope are at higher pressures, where retrieved values are less reliable. Similar plots for CH_4 and H_2O indicate consistent overestimates of descent above ~ 520 K, but CH_4 and H_2O do not suggest a substantial underestimate at lower levels.

Most of the ozone loss during the periods observed by MLS occurs in the late winter, February and March. Maps of MLS and calculated ozone for the north-looking periods in February/March 1993 and February 1996 (Figure 6) show the signatures of chemical ozone loss, with the calculations after ~ 30 days showing much higher vortex ozone than MLS. The main cause of higher extravortex ozone in the calculations at 520 and 585 K is the overestimate of de-

scient at these levels; higher values in 1996 suggest a larger overestimate in that year, but there are no contemporaneous long-lived trace gas observations with which to test this suggestion. Since vertical ozone gradients are much stronger outside than inside the vortex at these levels (e.g., Figure 2), a small overestimate of descent leads to a much larger discrepancy in transported values outside than inside the vortex.

Plots of ozone as a function of EqL and time for MLS and the LT model in February 1996 (Figure 7) show the rapidity and extent of ozone loss during that period. The difference between MLS and LT calculations gives an estimate of the chemical ozone loss. The substantial decrease outside the vortex (the black SPV contours demark the vortex edge region) at 585 K suggests an overestimate of descent, and thus the change in the vortex probably overestimates the chemical loss at this level. Ozone loss is more rapid near the vortex edge at each level. In February 1996, the lower stratospheric vortex was distorted by underlying tropospheric ridges, and the low temperature region was located near the vortex edge, in the region of strong winds [e.g., Manney *et al.*, 1996b, 2002]. Thus, not only was the activated chlorine transported most rapidly around the vortex edge region, but also the air in that region was exposed to more sunlight than if the vortex had been more pole centered; these conditions are consistent with more rapid ozone loss along the vortex edge.

Similar difference plots for 1992 through 1994 (MLS data for 1995, 1997 and 1998 are too sparse to make contour plots) at 465 K (Figure 8) show large interannual variability in ozone loss. As in 1996, ozone decreased more rapidly near the vortex edge in late February 1993; this was another period when the polar vortex was shifted off the pole and the cold region was often near the vortex edge [e.g., Waters *et al.*, 1995]. In contrast, in early March 1994, ozone decreased most rapidly near the vortex center; during this period, the low temperature region was more centered in the vortex, and the vortex itself was more pole-centered. Small deficits of observed versus model ozone outside the vortex in 1992 and 1993 suggest that the model may, during these periods, either overestimate descent even at 465 K, or underestimate transport of vortex air (which was higher in ozone than extravortex air at these latitudes during both these periods, despite substantial ozone loss in 1993) to midlatitudes; these two processes cannot readily be distinguished by examining long-lived trace gas data, as both too much descent and too little export of vortex air would result in lower (higher) N_2O and CH_4 (H_2O) values. The similarity in the magnitude of the model/data differences inside and outside the vortex in 1992 makes it difficult to unequivocally attribute the vortex difference to chemical loss, and, in fact, temperatures were not very low and chlorine was becom-

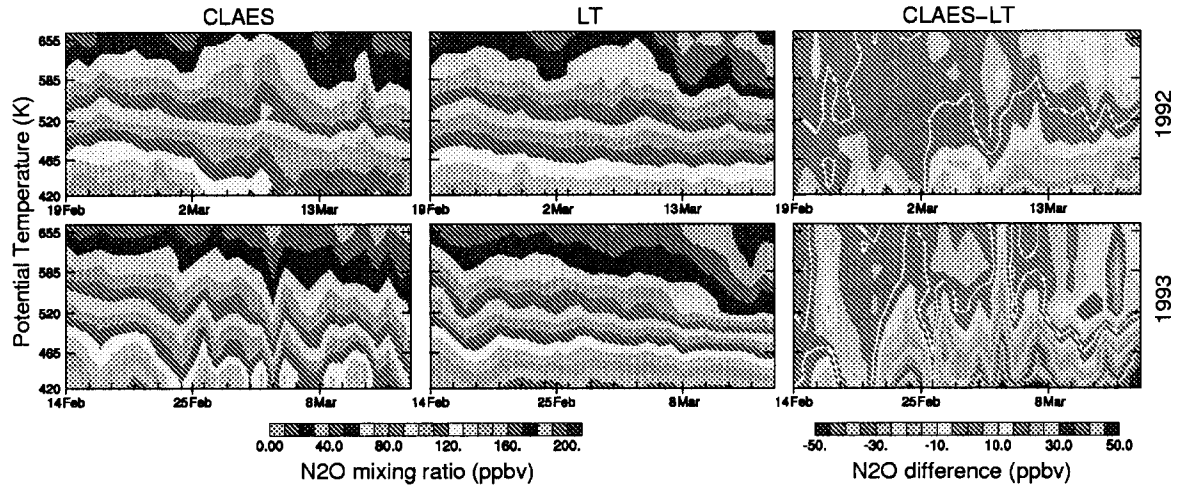


Figure 5. Vortex-averaged N_2O (ppbv) from CLAES (left), LT model (center), and the difference (CLAES-LT) between them (right), as a function of time and θ for the late winter observing periods, in (top, bottom) 1992 and 1993. N_2O is averaged within the $1.4 \times 10^{-4} \text{ sPV}$ contour (center contour in Figures 6–8). Missing days in CLAES plots are interpolated linearly from the values on the preceding and following days.

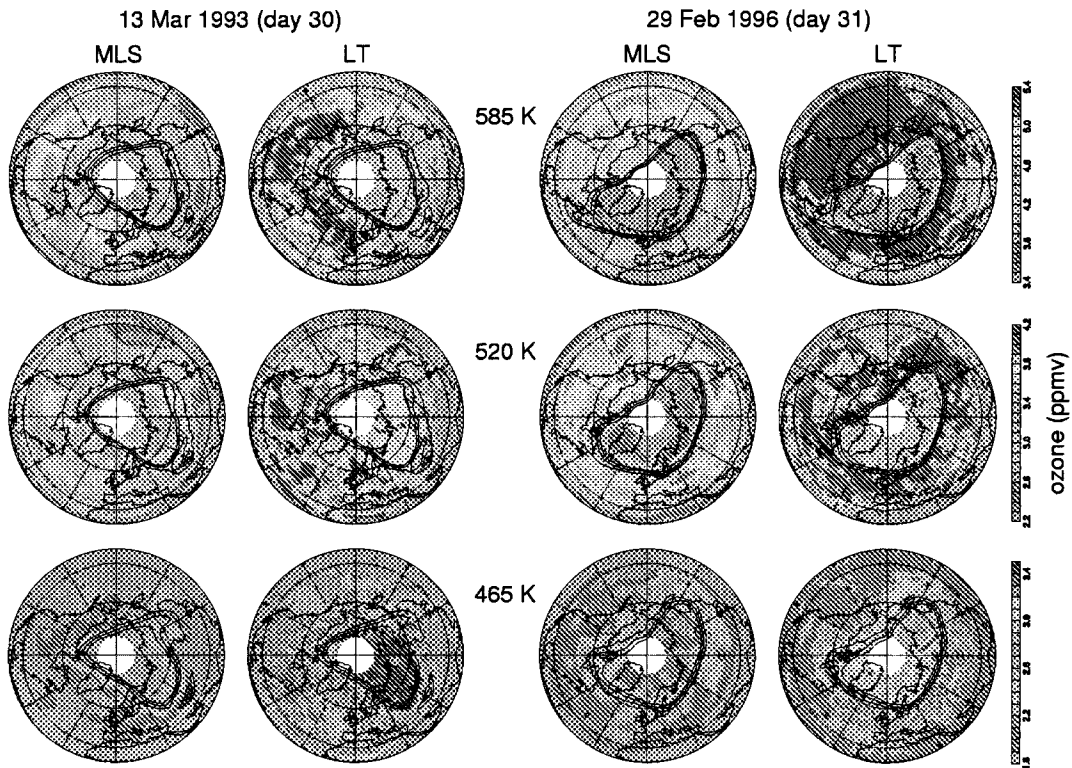


Figure 6. Maps of ozone (ppmv) from MLS data and Lagrangian trajectory (LT) model for late winter cases in 1993 (left two columns) and 1996 (right two columns) for day 30 (31) of the LT runs in 1993 (1996). Maps are shown for (top to bottom) 585, 520, and 465 K. Overlaid contours are scaled PV (sPV) of 1.2 , 1.4 and $1.6 \times 10^{-4} \text{ s}^{-1}$. Map projection is orthographic, with 0° longitude at the bottom and 90°E to the right, from 0 to 90°N, with dashed circles at 30°N and 60°N.

ing deactivated during this period [e.g., Waters et al., 1993, M. L. Santee, et al., “A climatology of lower stratospheric CIO based on UARS Microwave Limb Sounder measurements”, in preparation].

The time evolution of vortex-averaged ozone (Figure 9) shows the altitude distribution of ozone changes. The large decreases at and above 585 K in each of these plots reflect the overestimate of descent at these levels, and they suggest that the amount of that overestimation varies from year to year (e.g., it appears larger in 1994 and 1996 than in 1992 and 1993). A rough calculation, using the observed N_2O vertical gradients (e.g., Figure 5), observed and LT model N_2O time evolution, and observed vertical ozone gradients, indicates that the amount of the overestimate of descent at 585 K could result in an erroneous estimate of the increase in ozone at that level of $\sim 0.13\text{--}0.2$ ppmv at the end of the February 1992 and February 1993 periods (with values toward the larger end of this range for 1992 and the smaller end for 1993); these values could account for all of the difference seen between the LT model and MLS ozone at this level in 1992 (suggesting no evidence of chemical loss at 585 K in February/March 1992) and $\sim 2/3$ of the difference in 1993 (suggesting a very small amount, $\lesssim 0.1$ ppmv, of chemical loss at this level in February/March 1993). Maximum ozone loss values were comparable in 1993 and 1996, but the region of large loss extended to ~ 550 K in 1996, compared to ~ 480 K in 1993. This is consistent with the greater vertical extent of chlorine activation seen in 1996 than in 1993 (M. L. Santee, et al., “A climatology of lower stratospheric CIO from UARS Microwave Limb Sounder measurements”, in preparation). Also, we are comparing mid-March 1993 with early March 1996; ozone loss in 1996 continued through March [e.g., Rex et al., 1997]. Considerable interannual variability in the vertical ozone gradients in the vortex – and interannual variability in the diabatic descent rates – leads to large interannual variability in the amount of replenishment of vortex ozone by descent, which masks chemical loss.

4. Quantification of Ozone Loss in the Lower Stratosphere

Figure 10 shows quantitative estimates of vortex-averaged ozone loss during the February/March Arctic observing periods of MLS in 1992 through 1998 (note that the observing periods begin and end a few days earlier each successive year, Figure 1). As shown in section 3, there is large interannual variability in Arctic ozone loss, both in observed ozone changes and in the amount of masking by descent in the vortex. Significant chemical ozone loss occurred during the late winter MLS observing periods in each year ex-

cept 1998 and possibly 1992. Largest loss was in 1996, with nearly as much in 1993 resulting from less observed ozone change, but greater modeled replenishment. Note, however, that if we compare the same dates (e.g., 1 March 1996 and 1993), there is only about $2/3$ the amount of ozone loss in 1993 as in 1996.

The 1997 calculations indicate only a relatively small amount of ozone loss, despite the prolonged cold in that winter [e.g., Coy et al., 1997]; this is partly because considerable additional ozone loss occurred after the end of the February MLS observing period [e.g., Manney et al., 1997; Schulz et al., 2000]. It may also result, in part, from the vortex and cold region being unusually pole-centered [e.g., Coy et al., 1997], so that air with activated chlorine may not have been spread as rapidly through the vortex [e.g., Manney et al., 2002]. Also, more ozone loss probably occurred north of 80°N (where MLS did not sample) after ~ 10 February (when this region began receiving sunlight); Schulz et al. [2000] showed that most ozone loss during this period occurred in the vortex core. In February/March 1992, temperatures near 465 K were above polar stratospheric cloud (PSC) formation thresholds, and MLS observed CIO only marginally higher than background values [e.g., Manney et al., 1994b, M. L. Santee, et al., “A climatology of lower stratospheric CIO from UARS Microwave Limb Sounder measurements”, in preparation]. Thus it is questionable whether the estimated chemical loss at this time is consistent with polar vortex conditions. As seen in Figure 8, there is a similar difference in ozone outside the vortex at this time, suggesting that there may be a bias in the transport calculation. Since the vortex was weak and highly variable at this time, transport of ozone poor air into the vortex could have resulted in a decrease that was not well-simulated by the model. Estimated ozone loss during the other periods appears qualitatively consistent with the temperatures and amounts of active chlorine (M. L. Santee, et al., “A climatology of lower stratospheric CIO based on UARS Microwave Limb Sounder measurements”, in preparation) in the vortex.

LT calculations for early winter (not shown) were also done for six of the seven years (excluding November/December 1997, when data extend only to 3 December), and from the end of each early winter north-looking period through the beginning of each late winter north-looking period (Figure 1). The early winter runs show evidence of a small amount of vortex-averaged ozone loss (~ 0.2 ppmv) at 465 K during 1–12 Jan 1992 and 22 December 1993–3 January 1994; in each of the other years, there is no evidence of significant ozone loss before mid to late January at 465 K. Calculations for the early winters of 1991–1992 through 1995–1996 all show $\sim 0.1\text{--}0.2$ ppmv ozone loss at

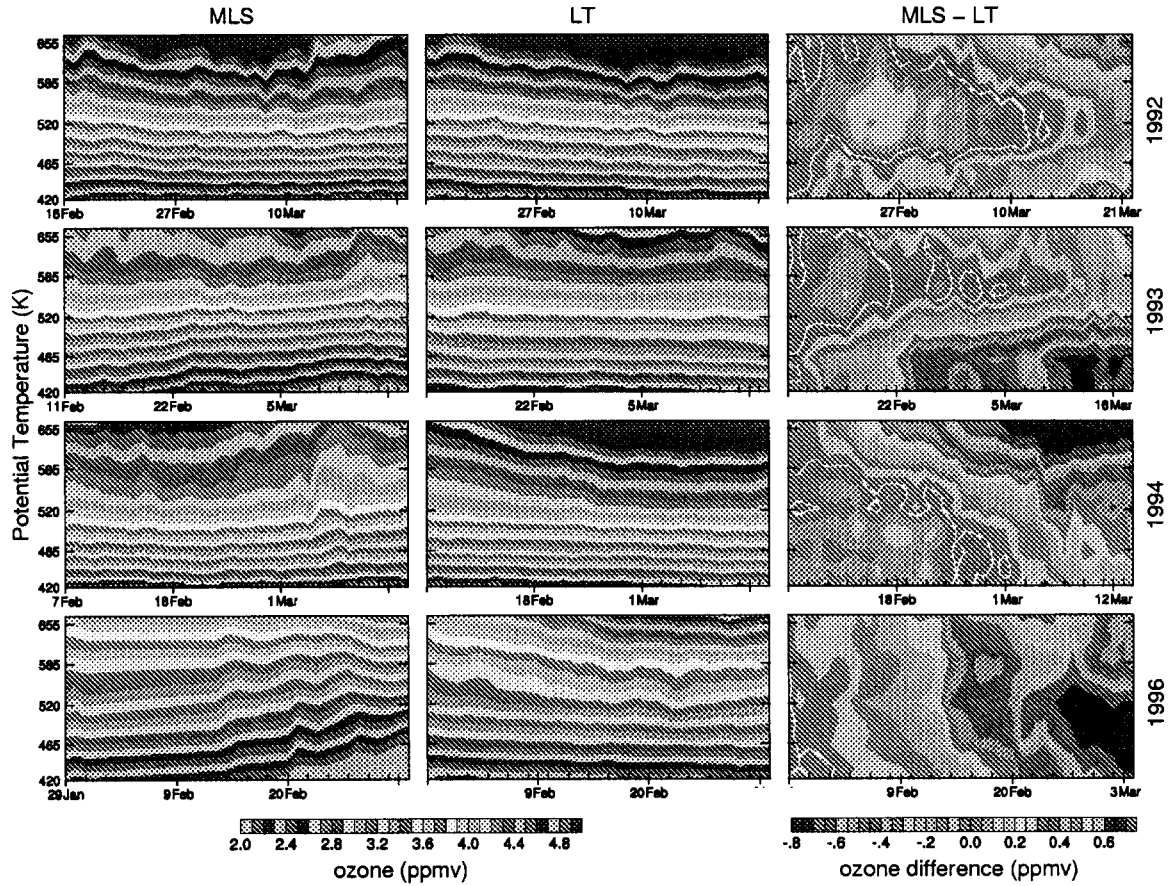


Figure 9. Vortex-averaged ozone (ppmv) from MLS (left), LT model (center), and the difference (MLS-LT) between them (right), as a function of time and θ for the MLS late winter observing periods, in (top to bottom) 1992, 1993, 1994, and 1996. Ozone is averaged within the $1.4 \times 10^{-4} \text{ s}^{-1}$ sPV contour (center contour in Figures 6-8).

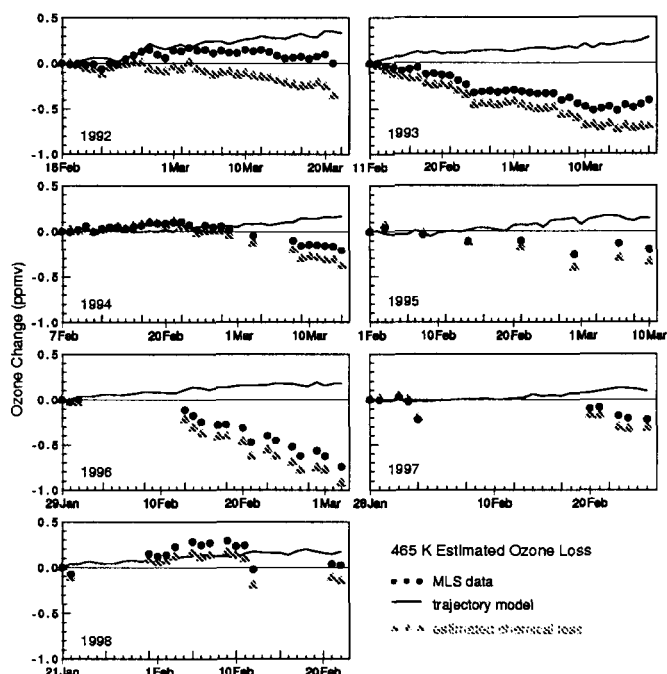


Figure 10. Estimates of vortex-averaged chemical ozone loss from MLS and the LT model at 465 K during the late winter observing periods in 1992 through 1998. Solid circles show MLS data averaged in the $1.4 \times 10^{-4} \text{ s}^{-1}$ contour; solid line shows LT model results; shaded triangles show the estimated chemical loss (MLS - LT model).

520 K, and ~ 0.3 – 0.5 ppmv at 585 K, with the smaller values in 1991–1992. No indication of loss was seen in December to January 1997, which was unusually warm. Each of the other years was cold in the early winter [e.g., Naujokat and Pawson, 1996; Zurek et al., 1996], and Goutail et al. [1999] have previously reported chemical loss in late December 1993. The long-lived tracer calculations indicate an overestimate of descent at 585 K in 1991–1992, but not in 1992–1993. Thus the decrease of ~ 0.3 ppmv at 585 K in 1991–1992 is probably an overestimate, but there is no evidence that the decrease of over 0.5 ppmv in 1992–1993 is.

Most of the midwinter runs indicate ozone loss during January. Calculations were reinitialized using the results of these runs as described in section 2 to estimate the amount of ozone depletion throughout the winter up to the end of each late winter observing period (these will be slight underestimates in 1992 and 1994, when there is evidence for a small amount of depletion before the beginning of the midwinter runs, and possibly at and above ~ 520 K, where there may also have been a little ozone depletion in early winter). The zero change for all curves in the extended run plots shown in Figure 11 is on the starting day of the midwinter run; hence, the difference on the first day shown indicates the ozone loss that occurred during the time MLS was observing the SH. The calculations for 1997–1998 are more uncertain than the others, because the gap between MLS observations was 50 days, well beyond the 20–40 days for which the LT calculations are expected to be reliable [e.g., Manney et al., 1995b, c, 1996b]. The low confidence in these LT model results is exacerbated by the occurrence of a strong warming in early January 1998 that severely eroded and distorted the vortex in the lower stratosphere; not only do the errors in the transport calculations grow faster under severely disturbed conditions, but also under such conditions the PV contour used for averaging may not be appropriate throughout the period. The MLS data may also be less robust during this period, as it was after a major change in instrument operations [e.g., Livesey et al., 2002]. Largest midwinter ozone loss at 465 K (about 0.6 ppmv) occurred in 1994–1995, which was unusually cold through late January [e.g., Naujokat and Pawson, 1996; Zurek et al., 1996]. ~ 0.3 – 0.4 ppmv midwinter loss was seen in 1991–1992, 1992–1993, 1995–1996, and 1997–1998. Temperatures were continuously below PSC formation thresholds through almost all of January 1992, 1993, and 1996 [e.g., Naujokat and Pawson, 1996; Zurek et al., 1996]. In 1994 and 1997, no evidence for ozone loss in the midwinter period is seen. Goutail et al. [1999] showed that ozone destruction in 1993–1994 occurred in late December and again in early March, consistent with our results and with the temperature evolution during that winter [e.g., Manney et al., 1995a]. In 1996–1997, while low tempera-

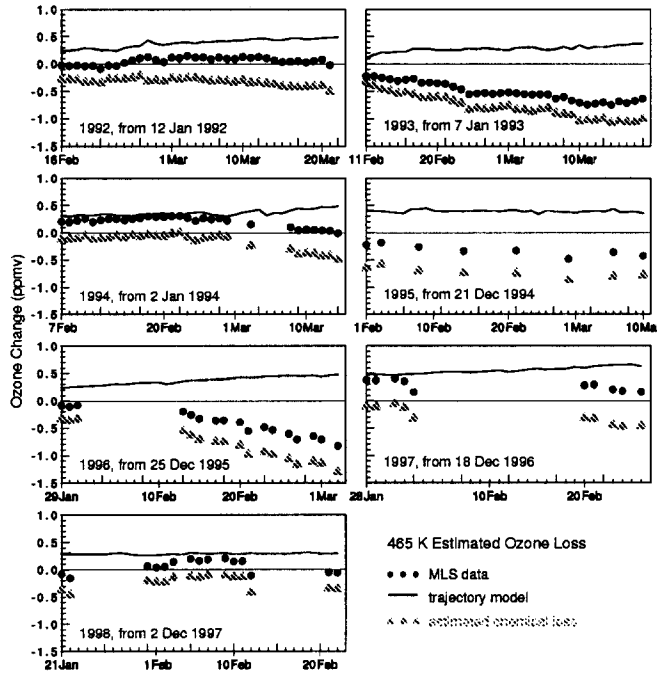


Figure 11. Estimates of vortex-averaged chemical ozone loss from MLS and the LT model at 465 K for “extended” (mid-to-late winter, see text) observing periods in 1991-1992 through 1997-1998. Layout is as in Figure 10.

Table 1. Estimated ozone loss at 465 K.

Dates	465 K Rate of Change ^a (ppmv/day)			Chem Loss ^b (ppmv)
	MLS	LT	Chem	
12 Jan–22 Mar 92	0.002	0.007	-0.005	-0.50
7 Jan–17 Mar 93	-0.013	0.005	-0.018	-1.01
2 Jan–14 Mar 94	-0.002	0.005	-0.007	-0.49
21 Dec 94–10 Mar 95	-0.006	0.004	-0.010	-0.78
25 Dec 95–3 Mar 96	-0.013	0.007	-0.020	-1.30
18 Dec 96–26 Feb 97	0.001	0.008	-0.007	-0.47
2 Dec 97–21 Feb 98	0.002	0.003	-0.001	-0.35

^a Calculated as the slope of a linear fit to the data, model and calculated loss curves in the above figures.

^b Total calculated change on the ending date of the LT model run; this gives the estimated chemical loss over the calculation period.

tures continued later than in any other year, the early winter (through mid-January) was unusually warm [e.g., Coy et al., 1997], so ozone loss was not expected until later.

Table 1 summarizes the estimated cumulative ozone loss from the extended runs (Figure 11). Comparisons with long-lived trace gases for 1992 and 1993 (section 3) indicated that the model does best near 465 K, with no evidence of a systematic bias in the masking by descent at these levels, so these are the values in which we have the greatest confidence. Table 2 gives the corresponding values at 520 K. Cal-

Table 2. Estimated ozone loss at 520 K.

Dates	520 K Rate of Change ^a (ppmv/day)			Chem Loss ^b (ppmv)
	MLS	LT	Chem	
12 Jan–22 Mar 92	0.005	0.008	-0.003	-0.28
7 Jan–17 Mar 93	-0.007	0.007	-0.014	-0.70
2 Jan–14 Mar 94	-0.002	0.010	-0.012	-0.66
21 Dec 94–10 Mar 95	0.0004	0.002	-0.002	-0.15
25 Dec 95–3 Mar 96	-0.013	0.010	-0.023	-1.35
18 Dec 96–26 Feb 97	-0.002	0.013	-0.015	-0.89
2 Dec 97–21 Feb 98	0.002	0.001	0.001	0.02

^a Calculated as the slope of a linear fit to the data, model and calculated loss curves in the above figures.

^b Total calculated change on the ending date of the LT model run; this gives the estimated chemical loss over the calculation period.

culations with the long-lived tracers suggest that the model sometimes slightly overestimates replenishment of ozone by dynamical processes at 520 K, but we still have fair confidence in these values. Values at higher levels are rendered highly uncertain by LT model overestimates of descent, and interannual variability therein. All years have significant ozone loss at 465 K, although, as discussed above, the value for 1998 is suspect; all years except 1998 show significant loss at 520 K. Again, 1993 and 1996 stand out with the most overall ozone loss. Both 1994 and 1997 show larger losses at 520 than at 465 K, in contrast to the other years.

The uncertainties in these estimates are very difficult to quantify. Uncertainties in vortex-averaged MLS ozone values (in particular for the differences that we are analyzing here, for which systematic errors can be ignored) are very small (less than ~ 0.03 ppmv), given the large number of profiles being averaged. The accuracy of the wind fields used in the LT calculations is problematic. Previous studies [e.g., Morris et al., 1995] indicated that uncertainties in the wind fields are a dominant effect in producing inaccurate trajectory calculations, and these effects are cumulative for the duration of the runs. Manney et al. [2002] showed very different results in isentropic trajectory calculations using different analyzed wind fields, and these differences varied from year to year with the meteorological conditions. The vertical velocities (in this case, diabatic descent rates from a radiation code) are particularly uncertain, and, for most of the years discussed here, we have no long-lived trace gas observations with which to test the transport. Manney et al. [1995b, c] suggested, based on sensitivity tests and long-lived trace gas calculations, that vortex-averaged ozone loss estimates from LT calculations at 465 K are expected to be reasonably accurate ($\lesssim 20\%$ uncertainty in the transport term) for 20–40 days, depending on the meteorological situation. We have done a number of additional sensitivity tests for the February/March 1993 case (shown to be one of the most sensitive to changing conditions) with the cur-

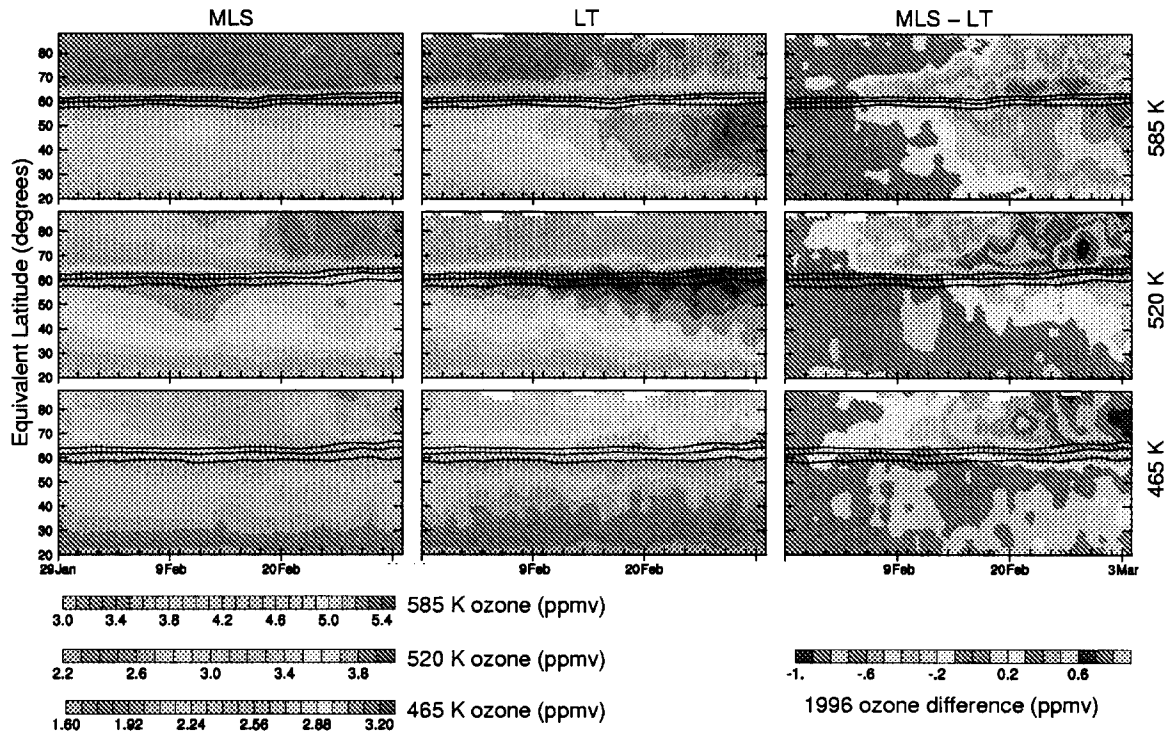


Figure 7. Ozone (ppmv) as a function of EqL and time from MLS (left), the LT model (center) and the difference (MLS-LT) between the two, for February 1996, at (top to bottom) 585, 520, and 465 K. Overlaid contours are $sPV = 1.2$, 1.4 and $1.6 \times 10^{-4} s^{-1}$. Missing days in MLS plots are interpolated linearly from the values on the preceding and following days.

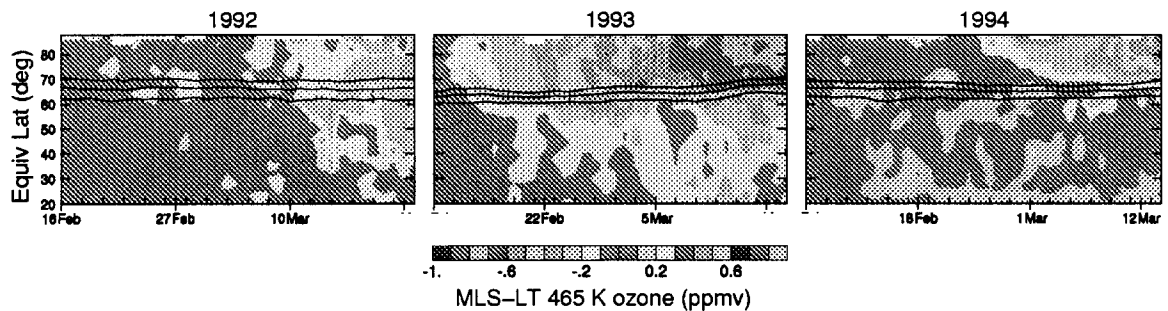


Figure 8. MLS-LT model ozone differences (ppmv) as a function of EqL and time during the late winter (February/March) observing periods in 1992, 1993 and 1994 (left to right), at 465 K. Layout is as in Figure 7.

Table 3. Estimated cumulative loss in column ozone above 100 hPa.

Time Period	$\Delta\text{Col100 (DU)}^a$	
	MLS	Chem
12 Jan–22 Mar 92	25	-29
7 Jan–17 Mar 93	27	-54
2 Jan–14 Mar 94	14	-35
21 Dec 94–10 Mar 95	18	-36
25 Dec 95–3 Mar 96	21	-63
18 Dec 96–26 Feb 97	11	-35
2 Dec 97–21 Feb 98	43	-22

^a Change in column ozone above 100 hPa between dates given in first column.

rent data and model configuration and confirmed these results. The uncertainties in the transport term at higher levels must be regarded as much larger, and typically biased toward overestimating ozone loss. Contributing further to the difficulty in quantifying the uncertainties in ozone loss estimates is the interannual variability in the importance of the transport term in the ozone loss calculation. Those years in which the transport term is small compared to the MLS observed changes will provide us with the best estimates of ozone loss; in other years, the uncertainties will be comparable to those in the transport term. Thus, our best guess is that estimates of cumulative loss at 465 K in 1992–1993, 1994–1995 and 1995–1996, and at 520 K in 1992–1993 and 1995–1996, are good to within 10–15%. Estimates in other years at 465 K (excepting 1997–1998) should be good to within 20–25%, and at 520 K to within ~ 30 –50%

5. Estimates of Column Ozone Loss

The amount of chemical loss of column ozone above 100 hPa (Col100) was estimated using Col100 calculated as described in section 2 for the late winter MLS observing periods (Figure 12), and for the extended runs to get an estimate of cumulative loss (Figure 13). Table 3 summarizes the estimated cumulative loss in Col100 on the ending date of the extended runs for each winter observed by MLS.

Largest column ozone loss was seen by MLS in early March 1996 and in mid-March 1993. Of the years observed by MLS, only 1996 and 1997 would be expected to show significant continuing loss beyond these last dates observed by MLS. The overestimate of ozone replenishment by descent above ~ 520 K in the LT model could result in a slight overestimate of the chemical loss in Col100; however, ozone mixing ratios at the levels where the overestimate is pronounced (at and above 585 K) do not contribute much to the column. Some calculations (e.g., Figure 5) suggest that the model may underestimate descent below ~ 450 K, but the long-lived trace gas data with which the descent rates are checked are not reliable enough below ~ 450 K to give con-

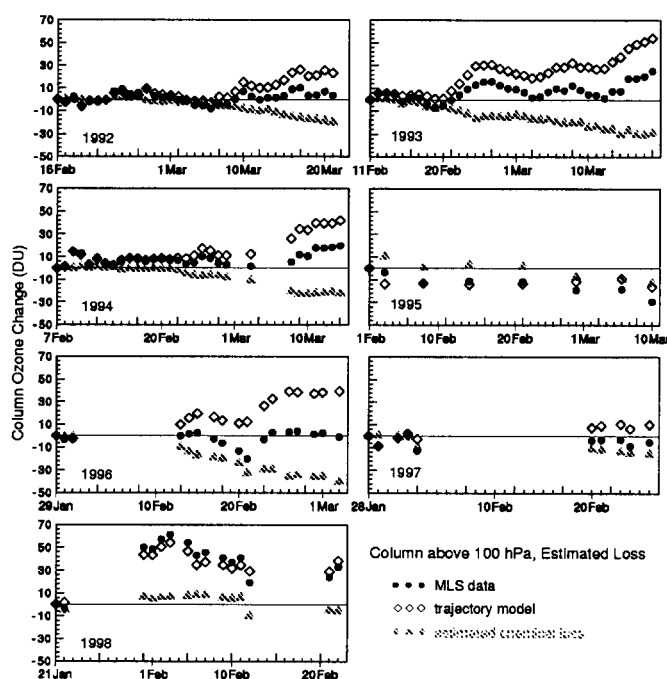


Figure 12. Estimates of vortex-averaged chemical loss in column ozone above 100 hPa (Col100) during the late winter observing periods in 1992 through 1998. Solid circles show MLS Col100 averaged in the $1.4 \times 10^{-4} \text{ s}^{-1}$ contour at 465 K; dark shaded diamonds show LT model Col100; light shaded triangles show the estimated chemical loss (MLS - LT model).

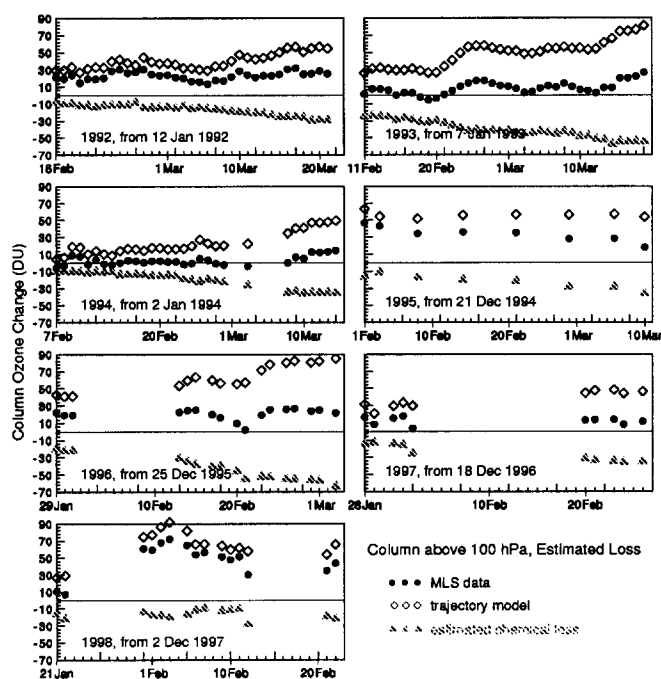


Figure 13. Estimates of vortex-averaged chemical loss in column above 100 hPa (Col100) during the “extended” (mid- to late-winter, see text) periods in 1991-1992 through 1997-1998. Layout is as in Figure 12.

fidence in this result. If the LT model does underestimate descent at lower levels, this might result in a more serious underestimate of the chemical change in Col100; however, descent rates at these levels are very small by any estimate, so masking by descent is a very small contribution to the ozone loss estimate. Thus, we believe that our Col100 loss estimates, like those for mixing ratio at 465 K, are typically uncertain by less than $\sim 20\%$.

Col100 loss estimated from MLS is much lower (often by up to $\sim 50\%$) than estimates of column loss in other studies [e.g., Müller *et al.*, 1997a; Goutail *et al.*, 1999; Harris *et al.*, 2002; Rex *et al.*, 2002, and references therein]. Goutail *et al.* [1999] showed significant ozone loss below 100 hPa during 1993-94 and 1994-95; Müller *et al.* [1997a] calculated column loss above ~ 150 hPa (in an isentropic layer from 350–550 K); Rex *et al.* [2002] calculated it in an isentropic layer from 400–700 K. Thus, the MLS limit at 100 hPa likely excludes significant ozone loss below this level. Another important consideration, especially in the later years, is that the estimates from MLS are for ending dates ranging from mid-March (1992) to late February (1998) (Figure 1), whereas estimates in most other studies are for near the end of March. 1995-96 [e.g., Rex *et al.*, 1997] and 1996-97 [e.g., Manney *et al.*, 1997; Sinnhuber *et al.*, 1998; Schulz *et al.*, 2000] had continuing chemical loss after the last MLS observing date in late winter. Assuming the loss rates during the late winter MLS observing periods continued until the end of March would result in ~ 33 DU and 19 DU more loss in 1996 and 1997, respectively. This would make our results for the 1996-1997 winter consistent within the uncertainties with Müller *et al.* [1997b] and Schulz *et al.* [2000]; our 1995-1996 results would then be ~ 70 -80% of the values obtained using ground-based observations and a transport model, and using HALOE tracer correlations [Harris *et al.*, 2002]. Differences in sampling also make comparisons of column ozone loss estimates particularly problematic, since ozone loss in an isentropic layer (as calculated in several of the studies mentioned here [Harris *et al.*, 2002]) may contribute more or less to column loss depending on the location of the sample (for warmer locations, the ozone mixing ratio changes on the isentropes will be at higher pressures, thus contributing more to chemical loss in the column). This may cause substantial biases in studies using sparser data, such as those from HALOE, POAM or ground-based observations, if measurements are concentrated on the warm or cold side of the vortex; these biases may vary from year to year as the position of the cold region and the observations inside the vortex vary.

6. MLS Observed Ozone Loss in 1999-2000

MLS made limited observations at high latitudes in early February and late March 2000 [Santee et al., 2000]. Since the observations did not provide complete coverage of the hemisphere, or even the polar regions, the LT method (which requires gridded hemispheric data for initialization) cannot be used in the same way described above. LT calculations were made for three periods during this winter by initializing with EqL/ θ "climatologies" based on MLS data. Among the "climatologies" considered were one based on MLS data surrounding the yaw dates in fall 1992 through fall 1993, one from LIMS observations in the 1978-1979 winter, ones based on MLS data from individual winters, and, the most complete one, based on averages throughout the winter of all the MLS V5 data during each 10-day period (referred to as MLSCLM). For calculations initialized on 2 February and 12 February 2000, a climatology was chosen based on its resemblance to the partial fields constructed from the MLS data on the initialization day, and calculations were also made using MLSCLM. For calculations initialized on 4 Jan 2000, no MLS data were available, and MLSCLM fields were used. Examination of EqL/ θ fields of MLS ozone over the years of observations indicates much less interannual variability in these fields in early January than by February.

The calculations for 2-12 February (Figure 14 shows those at 465 K using MLSCLM for initialization) suggest a rate of replenishment by diabatic descent of ~ 0.01 ppmv/day at 465 K, with similar values obtained for all initialization climatologies; with the decrease in MLS ozone of ~ 0.03 ppmv/day, we estimate a chemical loss rate during this period of $\sim 0.04 \pm 0.02$ ppmv/day. (Santee et al. [2000] showed a 0.04 ppmv/day decrease in vortex-averaged ozone from MLS; the difference between our estimate from MLS and theirs results from differences in the averaging: Santee et al. [2000] used unweighted averages of data at the MLS observation locations, whereas we used area-weighted averages of gridded data to produce results most comparable with those for the other years shown here; also, they used a less conservative definition of the vortex region.) The calculations suggest ~ 0.02 ppmv/day loss at 520 K and none at 585 K. During 12 February–29 March 2000 (not shown), calculations suggest loss rates (in ppmv/day) of about 0.006–0.012, 0.004–0.009, and 0.008–0.014 at 465, 520, and 585 K, respectively. These are broadly consistent with results from SOLVE/THESEO 2000 studies [e.g., Newman et al., 2002]. Results for 4 January–2 February 2000 are inconclusive given the uncertainty in the appropriateness of the initialization fields, but suggest substantial loss near 585 K (~ 0.015 – 0.030 ppmv/day) and show no evidence for loss at lower levels.

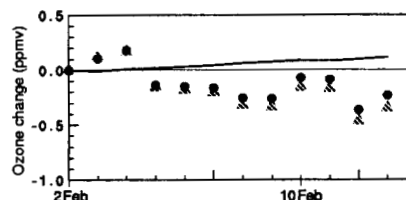


Figure 14. Estimates of vortex-averaged chemical ozone loss from MLS and the LT model at 465 K during 2-12 February 2000. Layout is as in Figure 10.

7. Summary and Conclusions

We have updated estimates of Arctic ozone depletion based on MLS data and Lagrangian transport (LT) calculations with V5, the definitive version of MLS data. The calculations have also been extended to all winters with sufficient MLS data, 1991-1992 through 1997-1998, and throughout those winters to provide estimates of cumulative ozone loss in each winter. LT calculations with long-lived trace gases – N_2O and CH_4 from CLAES and H_2O from MLS – during the 1991-1992 and 1992-1993 winters have been updated with current data versions to validate the LT model. LT calculations were also modified to produce rough estimates of ozone loss from the limited MLS data in the 1999-2000 winter.

The results show large interannual variability in the amount, timing, and patterns of ozone loss observed by MLS, consistent with the temperature evolution and structure in the different winters. In February/March 1993 and February 1996, when a large cold region was usually situated near the vortex edge and the vortex was shifted far off the pole, ozone loss was most rapid near the vortex edge. In contrast, in February/early March 1994, when the vortex and cold region were pole-centered, most ozone loss was seen near the vortex center. The greatest vertical extent of ozone loss seen by MLS was in early March 1996; the magnitude of ozone loss near 465 K at this time was similar to that in mid-March 1993, but ozone loss in 1996 both extended higher and continued longer than in 1993.

Calculations for the mid-winter periods (ranging from mid-January to mid-February in the earlier years to late-December through late-January in the later years), when MLS did not observe the NH, show evidence of significant ozone loss, except in January 1994 and late-December 1996 to late-January 1997, when it was too warm for PSC formation. At 465 K (near 50 hPa in cold periods), where we have the most confidence in our estimates, the loss during this period ranges from ~ 0.3 to 0.6 ppmv. In 1994-1995, most of the ozone loss observed by MLS occurred between 21 December 1994 and 1 February 1995. In early Janu-

ary 1992 and in late-December 1993 to early January 1994, there is evidence of ~ 0.2 ppmv vortex-averaged ozone loss at 465 K, consistent with previous studies [e.g., Rex et al., 1998; Goutail et al., 1999]. The LT calculations coupled with MLS observations suggest some ozone loss at 520 and 585 K in early to mid-winter in other years, but overestimates of replenishment by descent at these levels cause us to view these results skeptically. The exception is in early January 1992, when the LT calculations suggest ~ 0.6 ppmv loss at 585 K, and calculations with long-lived trace gases do not suggest an overestimate of descent at this time and level. It is of particular interest to quantify ozone loss in early to mid-winter, as modeling studies have not been successful in reproducing ozone loss at this time [e.g., Becker et al., 2000, and references therein].

The most severe Arctic ozone depletion seen by MLS was in 1995–1996, with ~ 1.3 ppmv accumulated chemical loss at 465 K for the winter by 3 March and in 1992–1993 (~ 1.0 ppmv loss by 17 March). Calculations of depletion in column ozone above 100 hPa indicate ~ 63 and 54 DU column loss on 3 March 1996 and 17 March 1993, respectively. The values for column loss are considerably smaller than those reported in the literature using other datasets and/or methods [e.g., Müller et al., 1997a; Goutail et al., 1999; Harris et al., 2002]. Two major reasons for this are: (1) the MLS estimates are for an earlier time period than other studies (limited by the period of MLS north-looking observations), which usually give estimates for near the end of March [e.g., Harris et al., 2002]; and (2) column ozone from MLS can be calculated only above 100 hPa; Goutail et al. [1999] showed significant ozone loss below this level, and this loss at higher pressures would contribute strongly to column loss.

MLS data in February and March 2000 were limited in both spatial and temporal coverage. A version of the LT model initialized with EQL/θ-space “climatologies” gives us a rough estimate of the ozone loss consistent with MLS observations. Ozone loss occurred at a rate of ~ 0.04 ppmv/day during 2–12 February 2000 and 0.006–0.012 ppmv/day during 12 February–29 March, and there was no evidence of chemical loss at 465 K during 4 January–2 February (although 0.015–0.03 ppmv/day loss was estimated at 585 K). These results are broadly consistent with other studies of the 1999–2000 winter [e.g., Newman et al., 2002].

The uncertainties in the LT calculations, and hence in our ozone loss estimates, are large and difficult to quantify. Uncertainties stemming from the analyzed wind and temperature fields used are common to any procedure for estimating ozone loss that involves a transport model and/or vertical velocities calculated from a model, such as the Match technique (although these uncertainties are reduced in the Match

method due to the short period of the transport calculations) [Rex et al., 1997, and references therein], the transport model based methods of Deniel et al. [1998], Goutail et al. [1999], Guirlet et al. [2000], Hoppel et al. [2002], and others, and “vortex-averaged” methods such as used by Knudsen et al. [1998]. Other methods, such as tracer correlations [e.g., Müller et al., 1997a, b, and references therein] have their own, correspondingly large, uncertainties [e.g., Michelsen et al., 1998; Plumb et al., 2000]. The uncertainties in all techniques, and the different sampling (both in vertical and horizontal coverage) and temporal coverage make it difficult to compare various methods in any great detail. The results shown here are not only more complete, but show some quantitative changes from those for MLS used by Harris et al. [2002]. Still, they generally confirm the conclusion of Harris et al. [2002] of broad overall agreement between many studies in the lower stratosphere near 450–480 K.

Although assimilated wind fields are ever improving, a great difficulty in verifying transport model based techniques currently is the lack of extensive records of distributed fields of long-lived trace gas data with good precision and resolution with which to validate transport calculations. These would also be helpful in verifying both vortex-averaged approaches and tracer correlation methods (especially if multiple long-lived tracers were available). Data from an improved MLS instrument and the High Resolution Dynamics Limb Sounder on the EOS Aura platform, to be launched in 2004, will provide simultaneous of ozone and one or more long-lived trace gases; these data should substantially advance our ability to validate transport in the polar lower stratosphere for use in future studies of ozone loss.

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